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# AUTORADIOGRAPHY AS A SAFEGUARDS INSPECTION TECHNIQUE FOR UNIRRADIATED LWR FUEL ASSEMBLIES

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S. B. Brumbach and R. B. Perry




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Nondestructive Assay Section
Special Materials Division

May 1978

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# AUTORADIOGRAPHY AS A SAFEGUARDS INSPECTION TECHNIQUE FOR UNIRRADIATED LWR FUEL ASSEMBLIES

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## ABSTRACT

A nondestructive autoradiographic method is described which can provide a verification that rods in the interior of unirradiated LWR fuel assemblies contain low-enriched uranium. Sufficient absorber must be used to reduce contributions to image density by beta radiation from U-238 daughters. When appropriate absorbers are used, the density of the image of a uranium-containing fuel rod is proportional to the U-235 enrichment in that rod. Exposure times as short as 1.5 hours can be achieved by using fast film and intensifying screens. Methods are discussed for reducing contributions to the image density of any single rod from radiation produced by all other rods in the assembly. The technique is useful for detecting missing rods, dummy rods, and rods containing depleted uranium. These defects can be detected by visual inspection of the autoradiographs. In its present state of development, the technique is not sensitive enough to reliably detect the difference between the various U-235 enrichments encountered in current BWR fuel assemblies. Results are presented for field tests of the technique at BWR and PWR facilities.

## I. INTRODUCTION

Risks of nuclear-weapons proliferation exist whenever special nuclear materials (SNM) are transferred from a weapons-possessing state to a non-weapons-possessing state. This risk exists because of the possibility of diversion of SNM from a program such as power generation to a weapons program. In order to reduce the risk of diversion, safeguards have been established to assure that none of the SNM being transferred is diverted into a weapons program. An essential element of these safeguards is inspection by

international agencies to ensure that all SNM is accounted for.

Most of the SNM transferred to non-weapons-possessing states is in the form of low-enriched uranium in fuel assemblies for light-water reactors (LWR). Effective safeguards of the uranium in unirradiated assemblies requires an ability to verify the U-235 content of all the rods in an assembly. At present, there are several nondestructive assay techniques which can verify the U-235 content of isolated, individual LWR fuel rods or of the rods on the outer periphery of fuel assemblies. What is needed, however, is a nondestructive technique for verifying the U-235 content of rods in the interior of a fuel assembly. The technique should be easy to use by inspectors in the field, and should include easily portable equipment.

Two inspection techniques have been investigated for application to LWR fuel assemblies. One technique, presently under study at Los Alamos Scientific Laboratory, uses active neutron interrogation of a portion of a fuel assembly.[1] A second technique, which will be described in this report, is autoradiography.

Autoradiography has previously been used to study the distribution of radioactive elements in reactor fuel materials.[2,3] Recently, autoradiographic methods have been applied to inventory verifications of plutonium-containing fast critical assembly fuel,[4] and one study reported the examination of low-enriched uranium fuel rods.[5] The method is based on the ability of spontaneously emitted X- and gamma radiation from uranium-containing fuel elements to expose photographic film. This radiation can produce an image of a fuel element which is in close contact with the detecting film. The optical density of the resulting image will depend on the amount of radioactive material present. The image density can be used as a measure of the concentration of radioactive material, providing that exposure time, isotopic composition, absorber thickness, source-film geometry, and film-processing conditions are held constant.

The following sections will describe the methods used in applying autoradiography to the verification of U-235 content. Results will be presented for a laboratory model fuel assembly and for commercially produced assemblies

examined at operating power reactors.

## II. APPLICATIONS TO U-235: ISOLATED FUEL ELEMENTS

### A. Minimization of Exposure Time

The most serious difficulty in obtaining an autoradiographic image of a single, isolated U-235-containing fuel element is the exposure time required for useful image density. The methods used for obtaining images of plutonium-containing fuel elements required only 15- to 45-minute exposures, depending on the source-film distance and the absorber thickness.[4] If these same methods are used for low-enriched U-235-containing fuel elements, exposure times of several days are required to obtain equal image density for equal fissile content. Such long exposure times are obviously not useful in an inventory technique which must be applied by inspectors in the field. The exposure-time problem was also recognized in the previous autoradiographic study of U-235 enrichment.[5]

The problem of lengthy exposures is caused by the low gamma-ray emission rate for U-235. This difficulty can be overcome by using intensifying screens with high-speed films. Intensifying screens act as light amplifiers. The gamma rays incident upon the screens cause fluorescence in the screen's coating material. Each gamma ray can produce several secondary photons of lower energy than the incident gamma ray. The screens are then used with film having the greatest sensitivity for the lower-energy photons.

In order to select the film-screen combination for the minimum length of exposure, several series of exposures were made with a reference stainless steel-clad fuel rod which contained uranium enriched to 16% U-235. In these series of exposures, the relative speeds of various screens were determined. Table 1 gives the results of a survey of common intensifying screens used with Kodak Ortho-G medical X-ray film. In all cases, the radiation source was the reference 16% U-235 rod. Filtration was provided by 0.030 in. (0.076 cm; 1 in. = 2.54 cm) of stainless steel. The greater the relative speed shown in Table 1, the shorter the exposure. All film was processed in a Kodak X-omat-B automatic processor. All screen materials, ex-

cept the thalium (Tl)-doped CsI and the europium (Eu)-doped $CaF_2$, were commercially prepared as intensifying screens. The $CaF_2$(Eu) and the CsI(Tl) were discs 0.086 in. thick, and the fuel-rod images produced by these discs were poorly defined. As indicated in Table 1, the shortest exposure required to achieve a given image density was obtained with Kodak Lanex screens. The results of Table 1 were obtained with the use of a transmission spot densitometer.

Table 1. Relative speeds of intensifying screens with Ortho-G film. Higher speeds correspond to shorter exposures.

| Screen | Relative Speed |
|---|---|
| No screen | 0.02 |
| DuPont HL | 0.04 |
| ZnS | 0.05 |
| DuPont 1K | 0.05 |
| Radeline UD | 0.05 |
| $CaF_2$(Eu) (poor image) | 0.06 |
| Kyoko SMP-108 | 0.1 |
| Kyoko SMP-308 | 0.1 |
| Radeline TL ($CaWO_4$) | 0.1 |
| DuPont CB-2 | 0.1 |
| Radeline F | 0.1 |
| CsI(Tl) (poor image) | 0.2 |
| 3-M Alpha-8 | 0.6 |
| Kodak Lanex | 1 |

A similar series of exposures was used to evaluate several types of film. Lanex screens were used in all cases, and, again, the 16% U-235 fuel rod was used as a radiation source. The results are presented in Table 2. As indicated, optimum results were obtained with Kodak-XR medical X-ray film processed in an automatic, medical-film processor. Kodak Ortho-G film was subsequently used in most of the technique development and evaluation program

because of our ready access to an industrial processor.

Table 2. Relative speeds of films with Lanex screens. Higher speeds correspond to shorter exposures.

| Film | Relative Speed |
|---|---|
| Kodak Industrex AA | 0.02 |
| Kodak Royal X Pan | 0.2 |
| 3-M Trimax XM | 0.3 |
| Kodak Ortho G | 0.5 |
| Kodak XR* | 1 |

*Processed in medical-type automatic processor.

When comparing speeds of film or screens, it is important that measurements be made in a film-density region in which the exposure-density relationship is known. It is easiest to work in a density range where this relationship is linear. For Ortho-G film, this linear density range extends from typical base densities of 0.3 to density 1.5. At higher densities, increasing the exposure gives progressively smaller increases in density.

In addition to the film types in Table 2, Polaroid Type-57, 3000-speed film was evaluated. The ease of processing Polaroid film made it worthy of consideration. When Polaroid film was used with Lanex screens, the fuel-rod images which were obtained took about 1.5 times longer than was required for Ortho-G film. These images were of a poorer quality than those obtained with medical X-ray films. The Polaroid film had to be removed from its packet, inserted into a screen-containing cassette for exposure, and then reinserted into its packet for processing.

## B. Contributions from U-238

Both the U-238 and the U-235 present in typical low-enriched LWR fuel contribute to the radiation emitted by fuel elements. Consequently, both uranium isotopes contribute to the density of the autoradiographic image of a fuel element. The most intense gamma radiation from LWR fuel is the 185 KeV gamma of U-235. Other weaker U-235 gammas are also found at 205, 163, and 144 KeV. Decay daughters dominate the gamma-ray spectrum from U-238, with the most intense emissions at 766 and 1001 KeV, from Pa-234m. The largest contribution to the photon flux from both U-235 and U-238 were X-rays resulting from alpha decay. These X-rays are more intense from U-235 due to its shorter half-life. However, it is very important to note that for low-enriched uranium most of the total photon flux is contributed by the U-238.

In addition to gamma radiation, the Pa-234m daughter of U-238 also emits 2.29 MeV beta radiation which can penetrate common fuel-element cladding, and which can efficiently expose film. In order to separate the contributions from U-235 and U-238, a series of identical exposures was made with identical fuel rods containing fuel of various U-235 enrichments. The fuel rods were clad in 0.012-in. thick stainless steel, and the film was in a lighttight paper cassette. Figure 1 shows a plot of the optical density of the rod image on Ortho-G film as a function of U-235 enrichment. Exposure time was 25 hours, and no intensifying screens were used. The optical density has been corrected for the base film density. The Hurter and Driffield optical density units used throughout this report are defined as the negative logarithm of the fraction of light transmitted by the film. As indicated in Fig. 1, there is a slight decrease in image density with increasing U-235 enrichment. Under these conditions, the image-density dependence on U-235 enrichment is too small to be useful in an enrichment-verification method. If the slope of the line in Fig. 1 is due to the effect of beta radiation, one would expect to be able to reverse the effect by using additional beta-absorbing material between the fuel rod and the film. The ideal absorber has maximum mass density for high beta-stopping efficiency and low atomic number for minimum gamma absorption. Stainless steel was selected, and the results of using an additional 0.018 in. of stainless steel are shown in

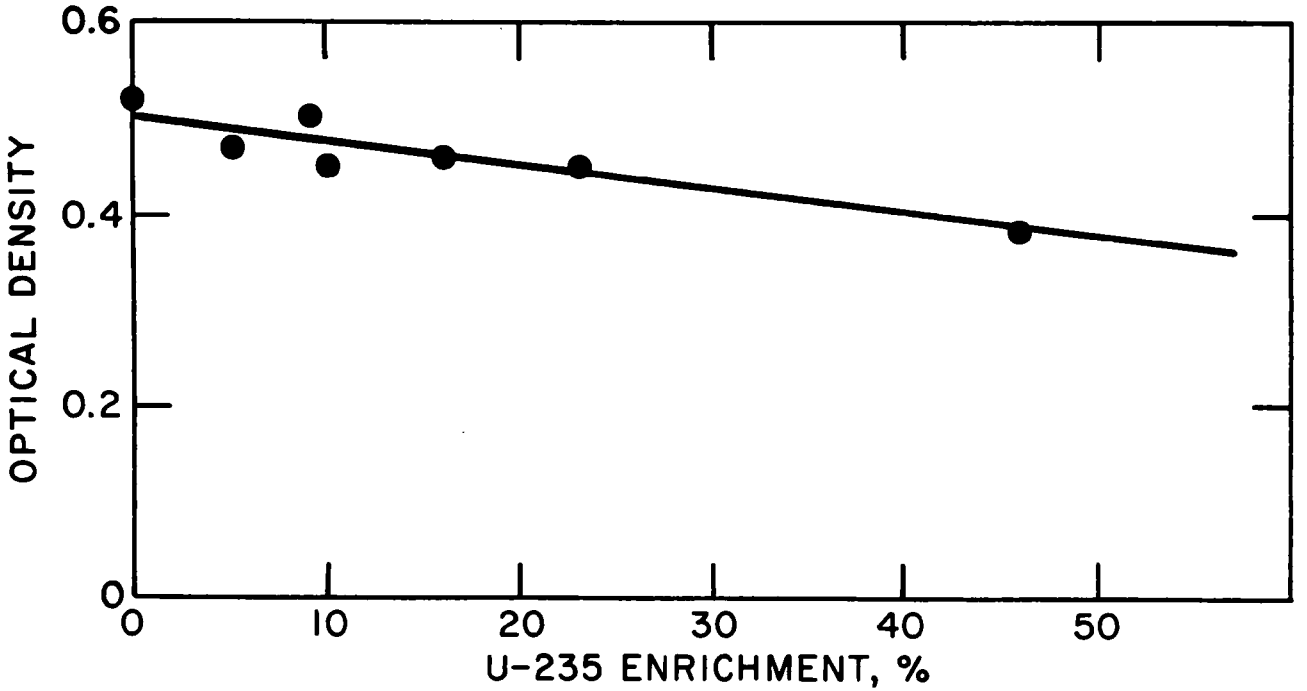



Fig. 1. Single-rod image density as a function of U-235 enrichment; Ortho-G film, no absorber, no intensifying screens. ANL Neg. No. 150-78-5.

Fig. 2. Though small, the slope of the line showing optical density versus the U-235 enrichment has changed sign compared to Fig. 1. A similar reversal of slope was also observed for Kodak-AA industrial X-ray film.

In order to increase the change in optical density of the autoradiographic image of a fuel element with U-235 enrichment, intensifying screens were added. The results for U-235 enrichment up to 23.3% are shown in Fig. 3. The 46% enrichment point was not included because it fell in the non-linear response range of the film. The results shown in Fig. 3 represent the greatest sensitivity to change in U-235 enrichment which was achieved in this study.

A positive slope of the line showing the image density versus the U-235 enrichment was also obtained when intensifying screens were used without additional beta absorber. This is still not sufficient protection against beta radiation effects, however, since a change in cladding thickness or cladding material can result in a greater image density for a given enrichment. As an example, autoradiographs were obtained for both aluminum-clad and stainless steel-clad fuel rods containing 3%-enriched uranium. When no additional beta absorber was used, the aluminum-clad rod produced an image of greater density than that produced by the stainless steel-clad rod. When an additional 0.018 in. of stainless steel absorber was used, the two image densities were equal. Even if the fuel rods under investigation in an inventory have normal cladding thickness sufficient to eliminate beta penetration, it is prudent to include extra beta absorber so that a potential diverter cannot mask reduced gamma activity by a tactic such as the substituting of depleted-uranium-containing rods which have thin or low-density cladding.

## III. APPLICATIONS TO FUEL ASSEMBLIES

### A. Model Assembly

In order to evaluate techniques for verifying the U-235 enrichment of fuel rods in assemblies, it was necessary to perform tests under fuel-assembly conditions which duplicated radiation background and spacing between rods. These conditions were duplicated in a specially constructed model of

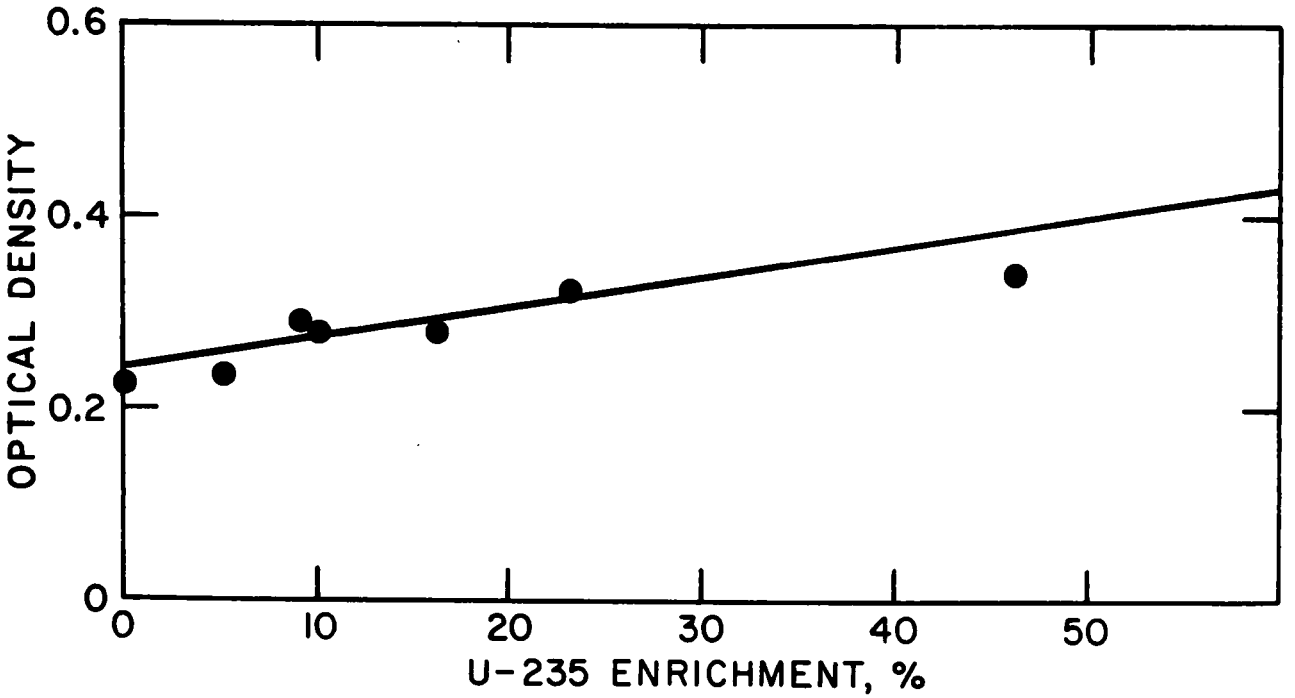



Fig. 2. Single-rod image density as a function of U-235 enrichment; Ortho-G film, 0.018-in. stainless steel absorber, no intensifying screens. ANL Neg. No. 150-78-6.

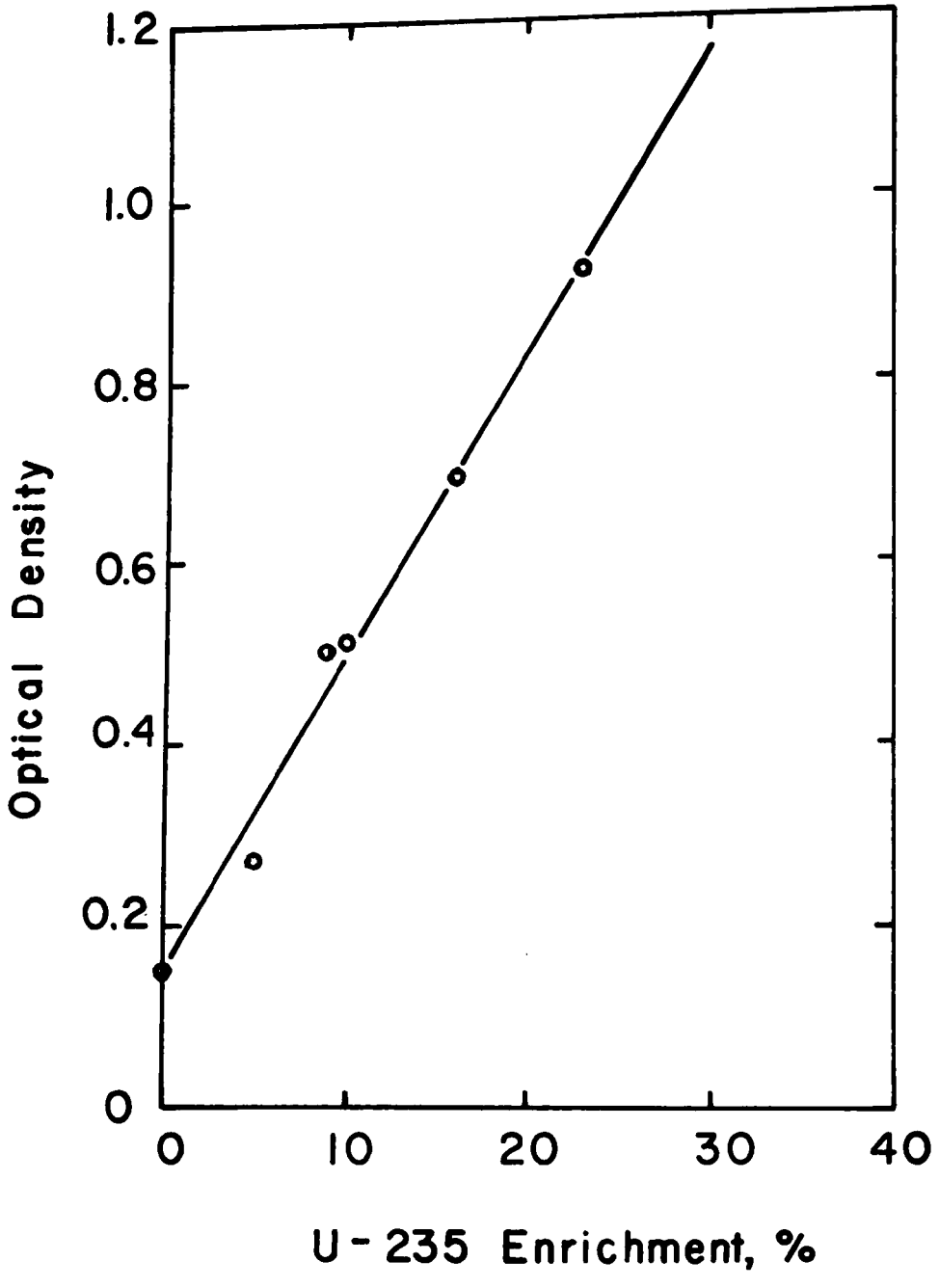



Fig. 3. Single-rod image density as a function of U-235 enrichment; Ortho-G film, Lanex screens, 0.018-in. stainless steel absorber. ANL Neg. No. 150-77-5.

a boiling-water reactor (BWR) assembly. The assembly consisted of a 7-by-7 array of rods. Each rod was approximately 51 in. long with an 0.416 in. OD, and was clad with 0.023-in. thick stainless steel. The rods were held in position by two 0.5-in. thick aluminum plates drilled with a square array of holes to accommodate the rods. The closest spacing between rods was 0.145 in., typical of BWR fuel assemblies. Each rod contained uranium enriched to 3.04% U-235. In order to test the detectability of rod substitution, solid steel rods and rods containing depleted uranium or 5%-enriched uranium were used. The depleted and the 5%-enriched uranium rods were only 6 in. long, and their stainless steel cladding was only 0.012 in. thick. Their OD was 0.375 in., but good rod-film cassette contact was maintained by wedging the special rods against the sides of the positioning holes facing the cassette.

## B. Internal Shielding: The Signal-to-Background Problem

### 1. Rods of Equal Enrichment

One major problem which limits the sensitivity of the autoradiographic technique to changes in U-235 enrichment is the high level of background radiation inside a fuel assembly. The density of the autoradiographic image of any single rod in an assembly receives a major contribution from all other rods in that assembly. In order to alleviate the problem, gamma-ray shielding can be used to isolate those rods which are under autoradiographic examination from all other rods in an assembly.

Film is most easily used to examine fuel rods in an assembly by the simple insertion of a film-containing envelope between two rows of rods. If no gamma absorbers are used, the film will record images of both rows of rods. In order to examine only one row at a time, a gamma absorber, such as lead foil, placed between the film and the row of rods not under examination will allow the row of the rods of interest to make a greater contribution to the final image density. For convenience, this lead foil can be included in the film cassette. Thus, each cassette will have a "front" side for exposure and a shielded "back" side. This gamma-ray shielding is, of course, not completely effective. Sixty-eight percent of the photons from

depleted uranium in the 66-283 KeV energy range were absorbed by 0.030 in. of lead. The fraction absorbed was 78% from a 5%-enriched uranium-containing rod.

This, however, still leaves the film open to radiation not only from those rods in the row of interest adjacent to the film, but also from all the rods on the unshielded "front" side. The amount of radiation from rods not in the row of interest, at the "front" side of the film, can be reduced by using a sheet of row-isolation shielding. This row-isolation shielding is placed parallel to the film, between the row of interest and the row of rods adjacent to the row of interest on the "front" side. The configuration of a film cassette and the row-isolation shielding in a model fuel assembly is illustrated in Fig. 4. The film cassette was a Kodak envelope for 70-mm wide X-ray roll film, and the lead shielding was made up of several 0.011-in. thick foils. Both the film cassette and the lead shielding were in polyethylene envelopes. The cassette and its constituents will be discussed in Section III-C.

The row-isolation shielding reduced the image-density contribution made by rods in all other rows. It did not, however, reduce the contribution to the density of a particular rod by those rods adjacent to it in the same row. In order to reduce this contribution from nearest neighbors within a row, it was necessary to remove the row-isolation shielding on the "front" side and to replace it with rod-isolation shielding which was inserted between the rows of rods perpendicular to the film. If a 7-by-7 array was to be examined, 6 pieces of rod-isolation shielding were needed instead of the single piece in the row-isolation case.

Any of the three types of internal shielding permitted the rods in the model assembly to be readily and unambiguously distinguished from one another under visual inspection of the film. Clear images were formed of all seven rods in any row. In all cases, the image density of the exterior rods was less than that of interior rods. The different methods of internal shielding did affect the contrast between the image density of the rods and the image density of the gap between the rods. A scanning densitometer was used to obtain image-density profiles from rows of rods in the

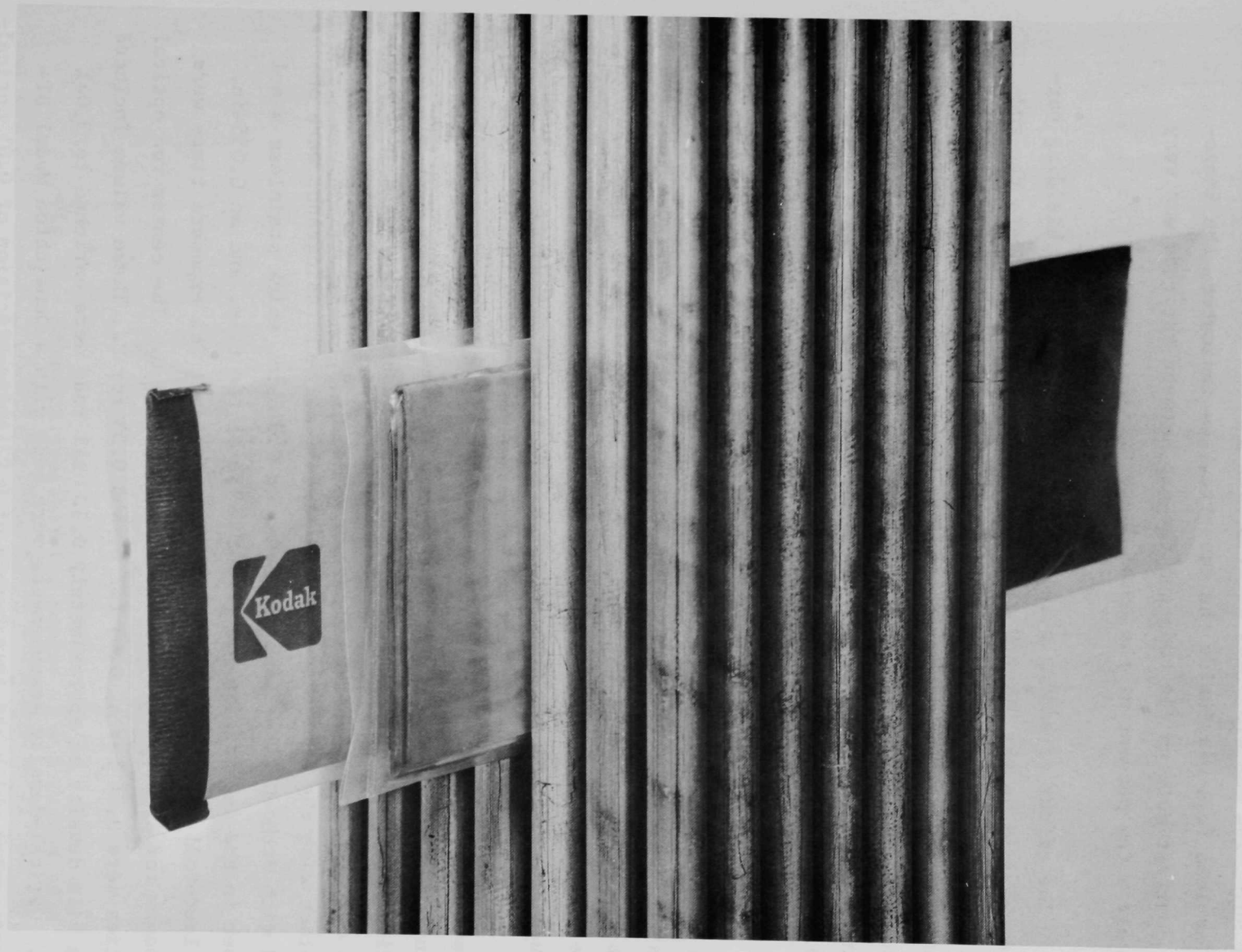



Fig. 4. Film cassette and row-isolation shielding in model assembly. ANL Neg. No. 150-78-2.

model assembly. The extent to which radiation from nearby rods contributed to the film density at and between rods is shown in Fig. 5-A, where the only internal shielding was 0.055 in. of lead foil on the "back" side of the film. The lead foil was inside the cassette. As indicated, the image-density contrast between the rods and the gap between the rods was very small—only a few percent of the total image density.

The effect of adding a sheet of lead row-isolation shielding parallel to the film on the "front" side is shown in Fig. 5-B. Here the rod-gap contrast had been improved so that the rods could be more clearly distinguished, although the density difference between rod and gap images was still no more than 10% of the rod-image density. Typical of the case in which two lead foils were parallel to the film, the two end rods are poorly defined.

The effect of using six pieces of rod-isolation shielding perpendicular to the film is illustrated in Fig. 5-C. This gave the best rod-gap contrast achieved in this study. Even with this optimum configuration, the rod-gap density difference was only about 25% of the total density. One advantage of the perpendicular lead configuration was that the end-rod images were much better defined. Whenever perpendicular lead shielding was used, it was important to keep the best possible contact between the lead foil edges and the film cassette. Nonuniform contact resulted in nonuniform rod-gap density differences for otherwise identical rods. In Fig. 5-C, the shielding foils were 0.125 in. thick. The model assembly was held in a horizontal orientation so that the weight of the lead shielding held the lead in contact with the film cassette. In Fig. 5, all autoradiographs were obtained with Ortho-G film, Lanex screens, an 0.018-in. thick stainless steel absorber in the cassette on the "front" side of the film, and an 0.055-in. thick lead-foil gamma absorber on the "back" side. The exposure times were 3.75 hours for A, 3.5 hours for B, and 4 hours for C. The center rod optical densities were 0.74 for A, 0.65 for B, and 0.53 for C. These values included a base film density of approximately 0.3. All rods were enriched to 3.04% U-235. Optical-density measurements were made with a Densichron Model DT-63 densitometer which had an accuracy of 0.02 and a precision of 0.01 optical-density units.

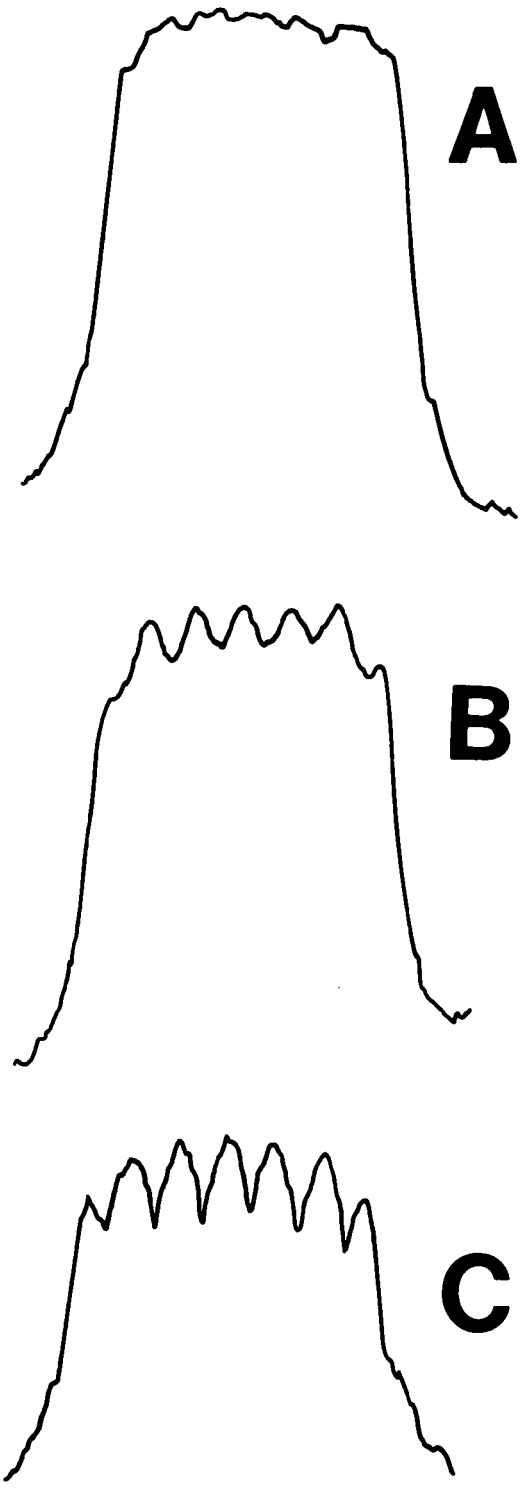



Fig. 5. Scanning densitometer traces of autoradiographs of a row of rods in the model assembly. A: No lead shielding on the "front" side; B: Row-isolation shielding; C: Rod-isolation shielding. ANL Neg. No. 150-78-17.

## 2. Detectability of Nonuniform Enrichment

Since the aim of this autoradiographic technique was to verify the U-235 content of rods in a fuel assembly, it was necessary to evaluate the technique for its ability to indicate deviations from expected or stated enrichments. The most elementary form of diversion from a fuel assembly is the removal of rods from the interior of the assembly and the leaving of void rod positions. In the model assembly, voids were easily detected by visual inspection of autoradiographs made with either row-isolation shielding parallel to the film, or rod-isolation shielding perpendicular to the film. The autoradiograph of a void in the center of a row of 3%-enriched rods with row-isolation shielding parallel to the film is shown in Fig. 6-A. If no lead shielding at all was used on the "front" side of the film, a void was still visually detectable, but the difference between the image of a void and the image of a 3%-enriched rod was subtle.

A slightly more sophisticated example of diversion might be the removal of U-235-containing rods and the substitution of "dummy" rods consisting of solid metal or metal tubes. When one 3%-enriched rod in the assembly was replaced by one solid steel rod of the same size, the substitution was readily detected by visual inspection of the autoradiographs. Detection was possible when row-isolation shielding and rod-isolation shielding were used, and was possible, though less obvious, when no lead shielding at all was used on the "front" side. An autoradiograph of a solid steel rod in the center of a row of 3%-enriched rods with row-isolation shielding parallel to the film on the "front" side is shown in Fig. 6-B.

At a somewhat higher level of sophistication, diversion could be attempted by substituting rods containing depleted uranium for rods containing enriched uranium. Such substitution of depleted-uranium rods was also detected by visual inspection of autoradiographs made when either row-isolation shielding or rod-isolation shielding was in place, and also when no lead was on the "front" side. Visual detection was easiest for the rod-isolation case, and most difficult when no lead at all was used on the "front" side. An autoradiograph of a depleted-uranium-containing rod in the center of a row of 3%-enriched rods with row-isolation shielding parallel to the

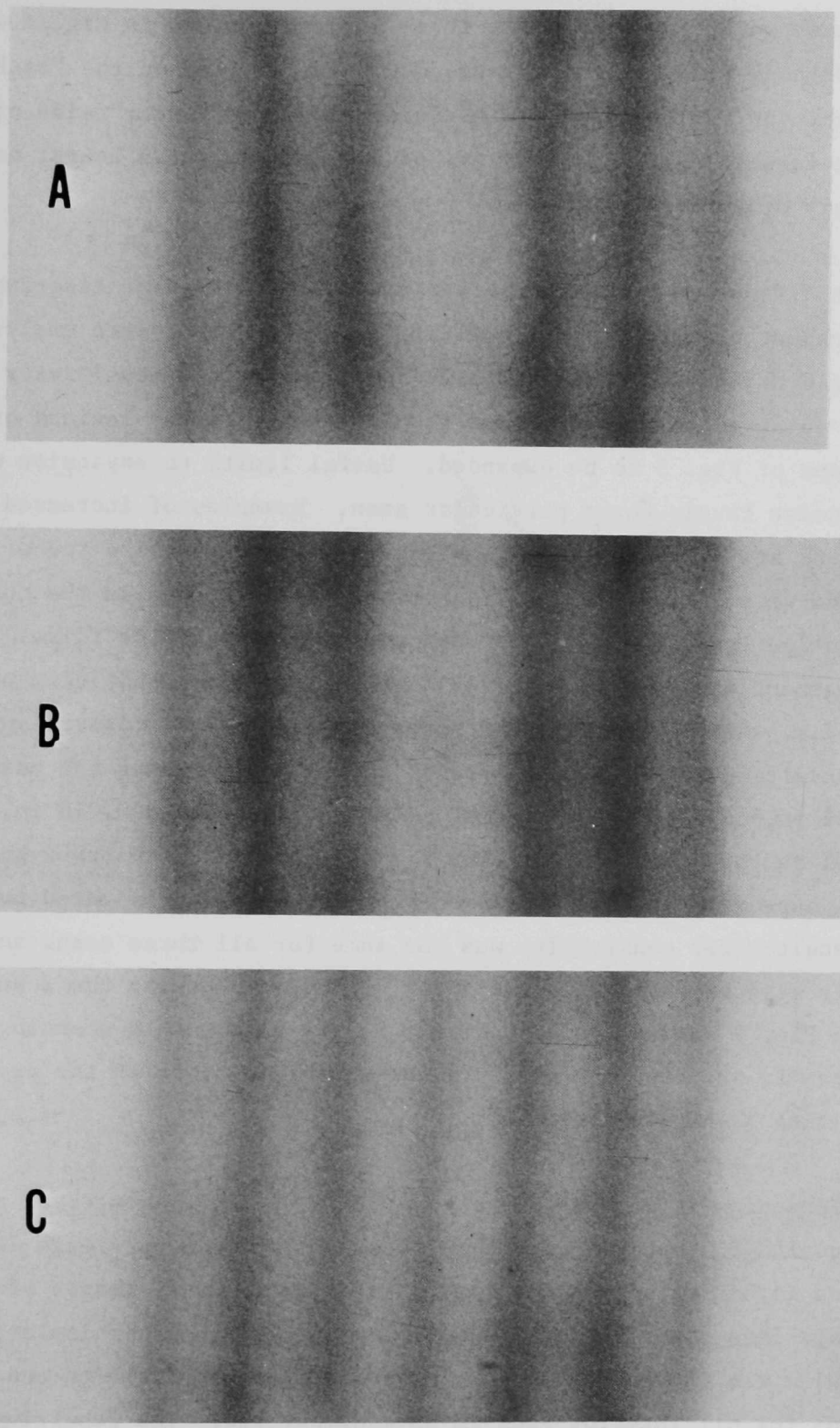



Fig. 6. Autoradiographs of a row of rods in the model assembly; Ortho-G film and Lanex screens. A: Center position void; B: Center position "dummy"; C: Center position depleted. ANL Neg. No. 150-78-19.

"front" side is shown in Fig. 6-C. All three autoradiographs in Fig. 6 were obtained with Ortho-G film, Lanex screens, 0.055 in. of lead on the "back" side of the film, and 0.018 in. of stainless steel on the "front" side of the film in the cassette; with an exposure of approximately 3.5 hours; and with the assembly oriented horizontally.

In an attempt to increase the sensitivity of the autoradiographic method to enrichment variation, the resulting autoradiographs were analyzed with a scanning densitometer. The scanning densitometer has sensitivity and zero-level adjustments which allowed the interesting upper regions of the density scans of Fig. 5 to be expanded. Useful limits to expansion were determined by noise levels for a particular scan. Examples of increased sensitivity scans are shown in Fig. 7. Figure 7-A is a scan of a row of 3%-enriched rods with a single depleted-uranium-containing rod in the center. Rod-isolation lead shielding was used perpendicular to the film. The same rod arrangement is shown in Fig. 7-B with row-isolation shielding parallel to the film. Similarly, Fig. 7-C shows the same rod configuration with no lead shielding at all on the "front" side. In all cases the cassette contained 0.055 in. of lead on the "back" side of the film and 0.018 in. of stainless steel on the "front" side. The film was Ortho-G with Lanex screens; exposures were approximately 3.5 hours; and the assembly was oriented horizontally. The densitometer sensitivity was the same for all three scans and was three times greater than the sensitivity used for recording the scans in Fig. 5. As Fig. 7 shows, analysis of autoradiographs with a scanning densitometer easily detected depleted-uranium-containing rods in the presence of rods containing 3%-enriched uranium.

The scanning-densitometer traces of Figs. 5 and 7 show that, even for rods of equal U-235 enrichment, there is some dependence of image density on rod position within an assembly. For constant enrichment, images of outer rods were always less dense than images of interior rods. A particularly difficult problem was the detecting of the substitution of depleted uranium-containing rods for <u>all</u> interior rods in an assembly row. The resulting density profile was slightly different when analyzed with a scanning densitometer. Although very subtle, the difference was also detectable by visual inspection. A spot densitometer, which measured absolute image density, was

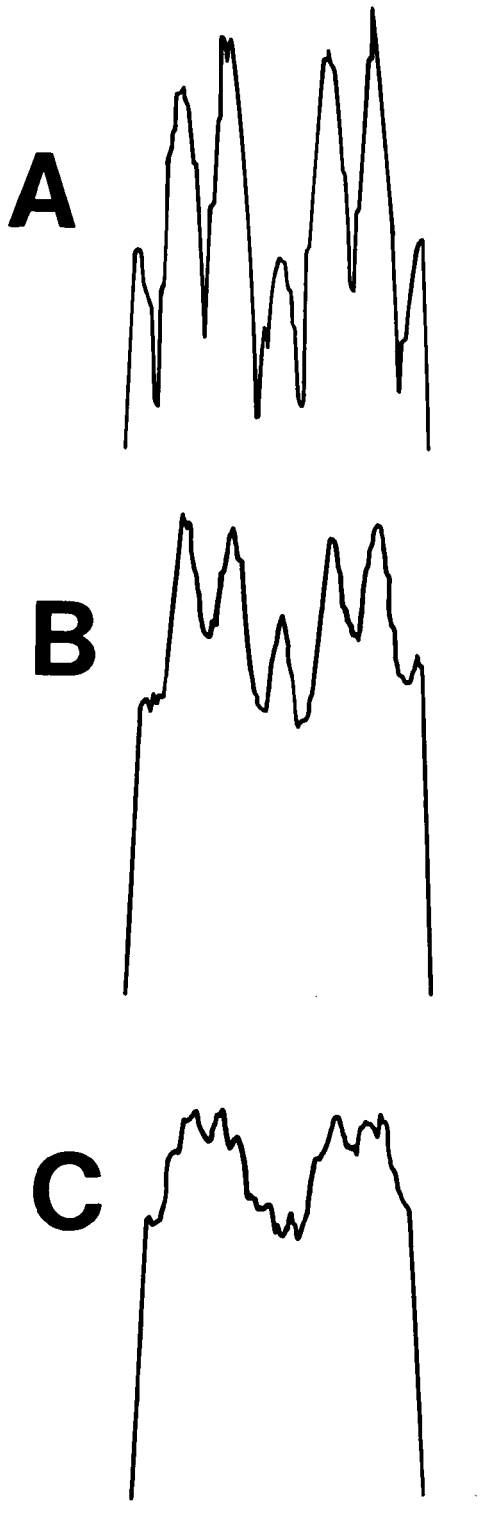



Fig. 7. Scanning-densitometer traces of autoradiographs of a row of rods in the model assembly. The central rod contains depleted uranium. A: Rod-isolation shielding; B: Row-isolation shielding; C: No shielding on the "front" side. ANL Neg. No. 150-78-18.

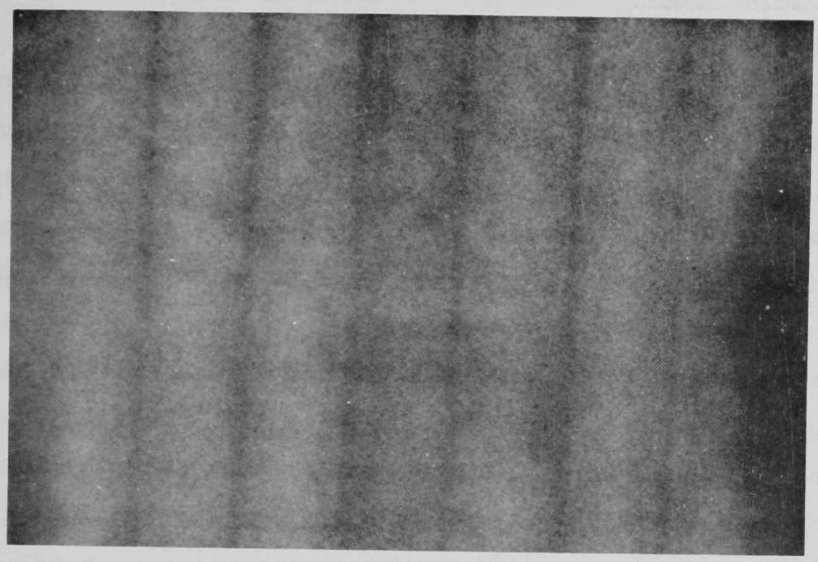
useful for interpreting these autoradiographs. The difference in density between the center rod and the end rod in a row was less when the interior rods contained depleted uranium than when they contained 3%-enriched uranium.

The depleted-uranium-containing rods used in the model assembly had thinner stainless steel cladding (0.012 in.) than the 3%-enriched uranium-containing rods (0.023 in.). In autoradiographs in which the 0.018-in. thick stainless steel beta absorber was not used, the images of the depleted-uranium-containing rods were more dense than the images of the enriched-uranium-containing rods. This provided still more evidence of the role played by beta radiation from U-238 daughters and of the need to include beta absorber material in film cassettes.

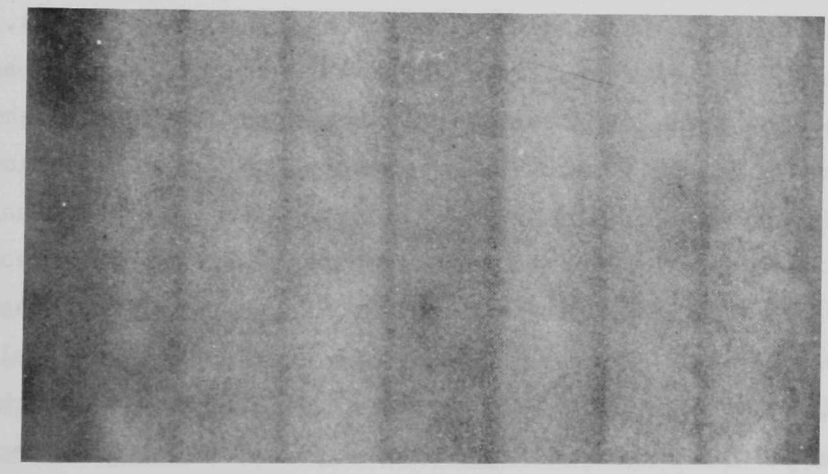
The only other low-enriched uranium-containing rod available for tests in the model assembly contained 5%-enriched material. The presence of a 5%-enriched rod in a row of 3%-enriched rods was readily detected by visual inspection. In order to evaluate the sensitivity of the method to smaller enrichment changes, it was necessary to use commercially produced BWR fuel assemblies. Results of these tests will be discussed in Section IV.

Several test exposures were made in the model assembly using Polaroid Type-57 film. Acceptable image densities were obtained in as little as 3.5 hours. The final image is a positive on paper, so the contrast is the reverse of that of the other films evaluated. Good quality images were obtained with Polaroid film when rod-isolation shielding perpendicular to the film was used, and the assembly was oriented horizontally. Figure 8 shows images obtained in the model assembly with rod-isolation shielding, a Lanex screen, stainless steel absorber, and the assembly in a horizontal orientation. In Fig. 8-A, the center rod contains depleted uranium, and, in Fig. 8-B, the center rod position is void. All other rods were 3% enriched. Exposures in which row-isolation shielding was used did not produce well-defined images of individual rods.

The normal exposure time for Ortho-G film in the model fuel assembly tests was 3-4 hours. As reported in Table 2, Kodak-XR film can be used to



A



B

Fig. 8. Autoradiographs of a row of rods in the model assembly; Polaroid film with Lanex screen and rod-isolation shielding. A: Center rod contains depleted uranium; B: Center position is void. ANL Neg. No. 150-78-21.

reduce exposure times even further. Some exposures were obtained with XR-type film, and images of ample density were obtained in 1.5 to 2 hours. This was the shortest exposure time used in this study for 3%-enriched material.

## C. Applications to Commercial Assemblies

The model fuel assembly described in Section III-A was useful in evaluating various techniques for inspecting commercially produced fuel assemblies. However, certain additional constraints must be considered before applying the methods described above to commercially produced assemblies. First, it must be recognized that fuel assemblies represent a substantial financial investment to a fabricator or to a power-generating utility. Because of this investment, great care must be exercised in the handling of any assembly for inspection purposes. One requirement is that any film cassette or shielding inserted into an assembly must contribute no chemical contamination to the fuel rods. This requirement is rather easily met by the placing of all materials in polyethylene envelopes prior to use.

A second requirement is that fuel rods in an assembly should not be stressed, bowed, or otherwise displaced. This, in turn, requires that cassettes or shielding packets be no thicker than the spacing between rods. This requirement, while not a serious problem for BWR assemblies, can be difficult to meet in some parts of pressurized water reactor (PWR) assemblies. The source of difficulty in PWR assemblies is the presence of guide thimbles for control rods which are spaced throughout the assemblies. In the case of the Westinghouse fuel assemblies for the Commonwealth Edison Zion reactors, the spacing between center positions of all rod types was 0.563 in. The outside diameter of the fuel rods was 0.422 in., which gave a fuel rod-fuel rod clearance of 0.141 in. This 0.141-in. spacing was sufficient for the placement of film cassettes and shielding. However, for most of the length of this assembly, the control-rod thimbles had an outside diameter of 0.546 in. The resulting clearance between a control-rod thimble and a fuel rod was only 0.079 inches. If both rows contained control-rod thimbles, the cassette had to be flexible enough to allow it to thread its way through the different 0.079-in. wide gaps. For part of the length of the assembly, the

control-rod thimbles were only 0.489 in. in diameter. Here, the clearance between a thimble-containing row and a row containing only fuel rods was 0.108 in.[6]

In normal power reactor fuel-handling and fuel-storage procedures, fuel assemblies are held in a vertical position. Thus, a means must be found for supporting the film cassettes and the shielding. The rod spacers, normally found at about two-foot intervals along the length of the assembly present the easiest source of support. However, if a section of the assembly not adjacent to a spacer is to be examined, or if heavy shielding is used, then another support mechanism must be found. The vertical orientation makes it difficult to hold rod-isolation shielding in place. For BWR assemblies, the lead must be held firmly against the film cassette by some external means. In the case of PWR assemblies, which typically contain 15-by-15 arrays of rods, the 14 pieces of lead shielding which would have been required were considered too unwieldy to be attempted.

The special requirements of commercial fuel assemblies led to a cassette composition which is illustrated schematically in Fig. 9. The beta absorber is in the cassette at the "front" side of the film, and the gamma absorber is at the "back" side. The film used in most tests was Kodak Ortho-G, and was 0.008-in. thick. The 0.016-in. thick fluorescent screens were Kodak Lanex and were made of plastic coated with La/Gd oxide. The beta absorber was stainless steel in all cases, 0.018-in. thick for the examining of the BWR assemblies, and 0.010-in. thick for the examining of the PWR assemblies. The thinner steel was used for the PWR assemblies because of the requirement for thin cassettes. The gamma absorber was made up of varying numbers of 0.011-in. thick lead foils. The lighttight envelope was either an 0.040-in. thick black plastic cassette or an 0.016-in. thick paper envelope used for packaging Kodak Ready-Pack X-ray roll or sheet film. The plastic cassettes were for either 5-by-7 in. or 8-by-10 in. sheets of film. The Kodak envelopes were 70-mm wide and of varying lengths, or were for 5-by-7 in. sheets. The 70-mm wide cassettes were much more convenient for use in commercial assemblies because of their light weight and easy placement. Size and weight could be reduced still further by using 35-mm wide cassettes. For the BWR assemblies, where thickness limitations were not severe, the lead shielding

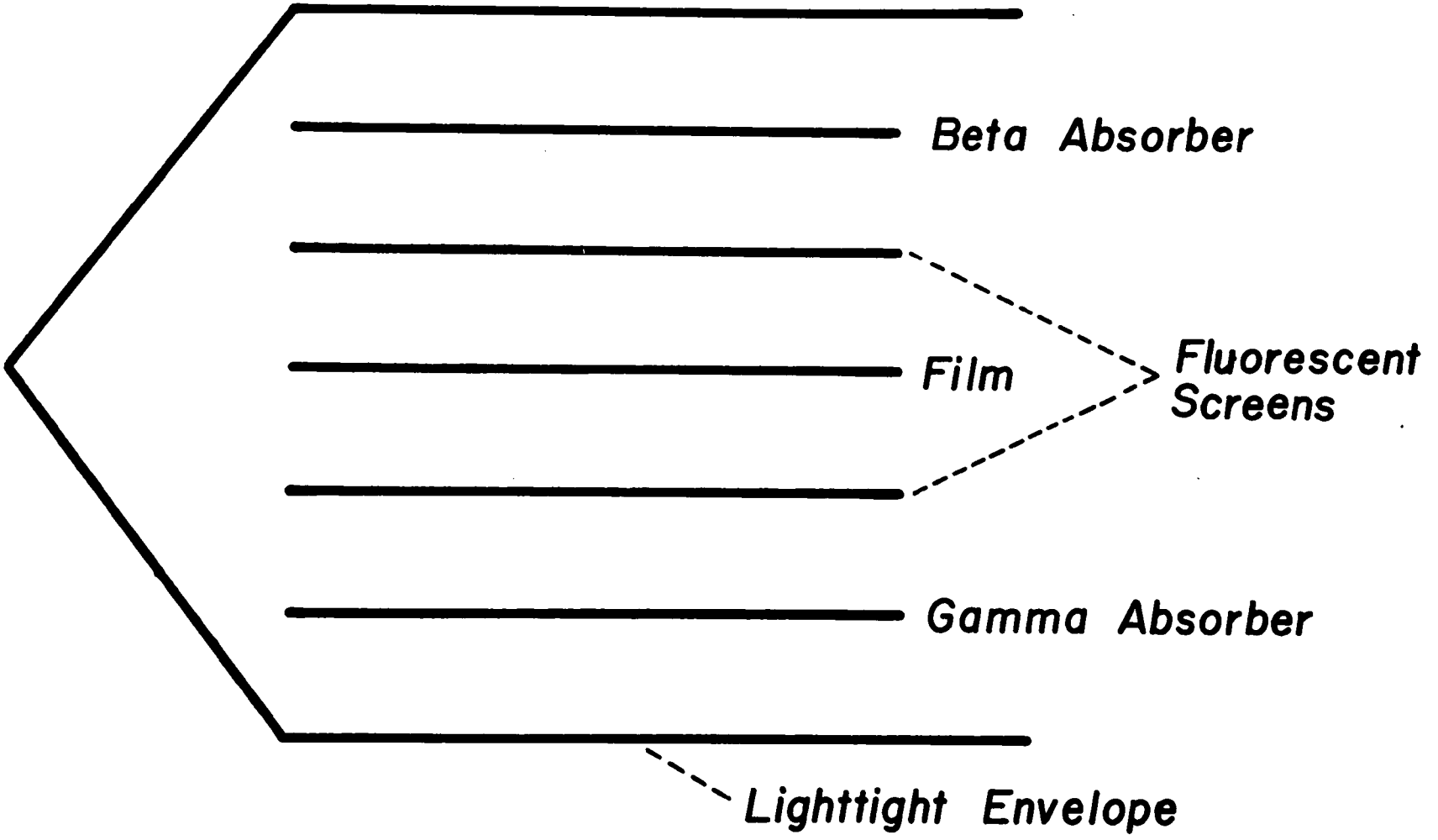



Fig. 9. Schematic diagram of the contents of a cassette for uranium autoradiography. ANL Neg. No. 150-77-14.

was an 0.125-in. thick sheet. When thickness limitations were severe, varying numbers of 0.011-in. thick foils were used. All cassettes and all shielding were finally enclosed in envelopes made of 0.006-in. thick polyethylene.

In some cases, Polaroid Type-57 film was used. In the darkroom, this film was removed from its packet and was inserted into a 5-by-7 in. plastic or paper Kodak cassette. Because the film had an opaque paper backing, only one screen was used. After exposure, the film was removed from the cassette and placed back in its packet for processing in a standard Polaroid film processor (camera back).

## IV. FIELD TEST RESULTS: COMMERCIAL FUEL ASSEMBLIES

### A. Boiling Water Reactor

Two separate field tests of the autoradiographic verification technique were conducted at the Dresden Station (BWR) of the Commonwealth Edison Company. The fuel assemblies for this reactor were 8-by-8 arrays of rods. Each rod had an 0.493-in. OD, and the gap between rods was 0.147 in. These assemblies were of particular interest because they contained rods of various U-235 enrichments in the range 1.45% to 2.87%. One rod, referred to as a water rod, contained no uranium at all. The autoradiographic tests at Dresden were conducted while the fuel assembly was on a test stand used for assembly inspection.

In the first test, the plan was to use the procedure which gave the best enrichment sensitivity, so that the potential sensitivity of the technique to enrichment change could be evaluated. Previous indications were that maximum sensitivity would be achieved with rod-isolation lead shielding perpendicular to the film. Four cassettes were prepared for the first test. Each contained 0.018-in. thick stainless steel beta absorber and 0.033-in. thick lead gamma absorber. Three cassettes contained Ortho-G film with Lanex screens, and one cassette contained Polaroid Type-57 film with one Lanex screen. Plastic cassettes were used in all four exposures. The lead shielding was 0.125-in. thick, and both the shielding and the cassettes were enclosed in polyethylene envelopes. Cassette thickness was approximately

0.143 in. Each cassette was placed between rows of fuel rods in different segments of the assembly as defined by rod spacers. The lead shielding was pressed against the film cassette by hand, and the cassette and the shielding were wrapped tightly in a polyethylene band which was, in turn, wrapped in tape to hold the shielding and the cassette in place. The film was exposed for approximately 4 hours and then was removed. After removal, all material which had been in contact with the assembly was checked for contamination and was subsequently removed from the reactor site.

When the cassette containing the exposed Polaroid film was opened, it was discovered that its chemical-containing pod had ruptured during handling, and this prevented any image formation. The three Ortho-G films were successfully developed and had formed images of fuel rods. However, the resulting autoradiographs all showed the common problem of poor, or nonuniform lead shielding-cassette contact during exposure. This resulted in good image separation between some rods and in poor separation between others. This nonuniform separation made analysis of the autoradiographs difficult. The autoradiograph of a row of rods which contained the water rod (no uranium) is shown in Fig. 10, along with a scanning-densitometer trace. The presence of the water rod (W) is obvious on both visual and densitometer-trace inspection. The rod at the far left of the autoradiograph is enriched to 2.14% U-235, while all other rods are enriched to 2.87%. This difference is not detectable by visual inspection, but the densitometer trace does indicate less density for the 2.14%-enriched rod on the left end than for the 2.87%-enriched rod on the right end. However, since other pairs of 2.87%-enriched rods in equivalent positions do not have equivalent density, one cannot conclude that enrichment-difference sensitivity is sufficient to distinguish between 2.14% and 2.87% U-235. The position of the water rod fixed the orientation of the autoradiograph with respect to the rods in the assembly.

Because of the many photographic steps required to produce positive images such as Fig. 10 from original autoradiographs, some variations in density contrast may occasionally exist between the positive images and the original negative autoradiographs. The image densities of the end rods in Fig. 10 appear to be such a variation.

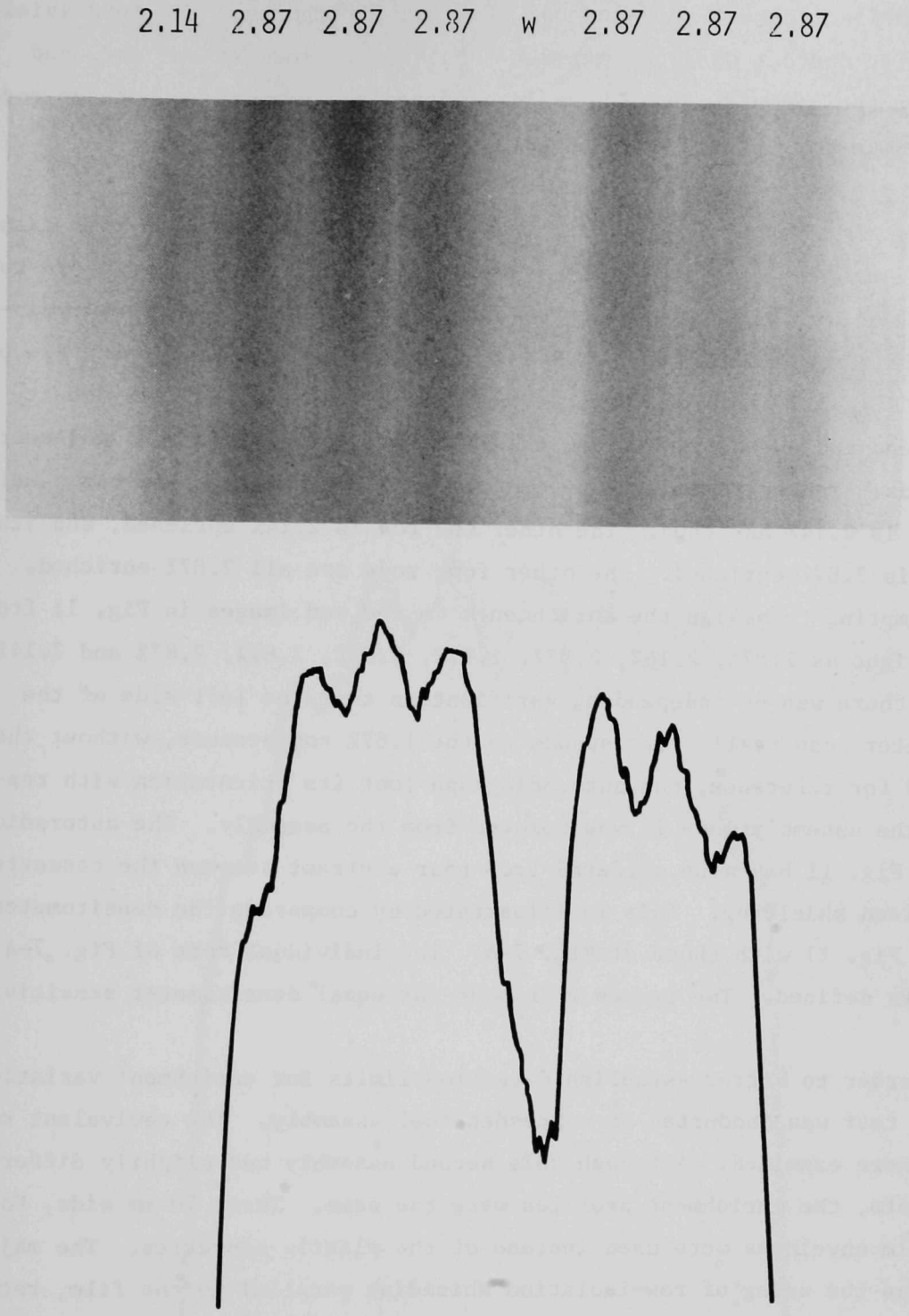



Fig. 10. Autoradiograph and scanning-densitometer trace of a row of rods in a BWR fuel assembly. Ortho-G film, Lanex screens, and rod-isolation shielding. Rod W contains no uranium. ANL Neg. No. 150-78-22.

The second autoradiograph examined a row of rods which contained seven rods of 2.87% enrichment, and one rod of 2.14% enrichment. The lead shielding-film cassette contact was poor between the last two rods at one end, and was good at the opposite end, which invalidated any comparison between rod-image densities at the two ends of the assembly.

In the third autoradiograph, the outside row of the assembly was examined. This row contained the largest variation in enrichment, with one rod of 1.87%, two rods of 2.14%, and five rods of 2.87% U-235. The autoradiograph of this row, and the corresponding scanning-densitometer trace are presented in Fig. 11. The image of the end rod on the left has less density than the end rod on the right, and this is also true for the rods adjacent to the ends. In the assembly, one end rod is 1.87% enriched and has a neighbor which is 2.14% enriched. The other end rod is 2.14% enriched, and its neighbor is 2.87% enriched. The other four rods are all 2.87% enriched. It was tempting to assign the enrichments to the rod images in Fig. 11 from left to right as 1.87%, 2.14%, 2.87%, 2.87%, 2.87%, 2.87%, 2.87% and 2.14%. However, there was no independent verification that the left side of the densitometer scan really corresponds to the 1.87% rod because, without the water rod for reference, the autoradiograph lost its orientation with respect to the assembly once it was removed from the assembly. The autoradiograph in Fig. 11 has also suffered from poor contrast between the cassette and the lead shielding. This is illustrated by comparing the densitometer scans of Fig. 11 with those of Fig. 7-A. The individual rods of Fig. 7-A are better defined. The traces were made for equal densitometer sensitivity.

In order to better establish detection limits for enrichment variation, a second test was conducted on a Dresden fuel assembly. The equivalent rows of rods were examined. Although this second assembly had slightly different enrichments, the enrichment profiles were the same. Three 70-mm wide, Kodak X-ray film envelopes were used instead of the plastic cassettes. The major change was the using of row-isolation shielding parallel to the film, rather than the perpendicular rod-isolation shielding which was used in the first test. This shielding change resulted in a much easier setup for exposure, and eliminated the shielding-cassette contact problem. Four exposures were made, including one with a plastic cassette containing Polaroid Type-57 film.

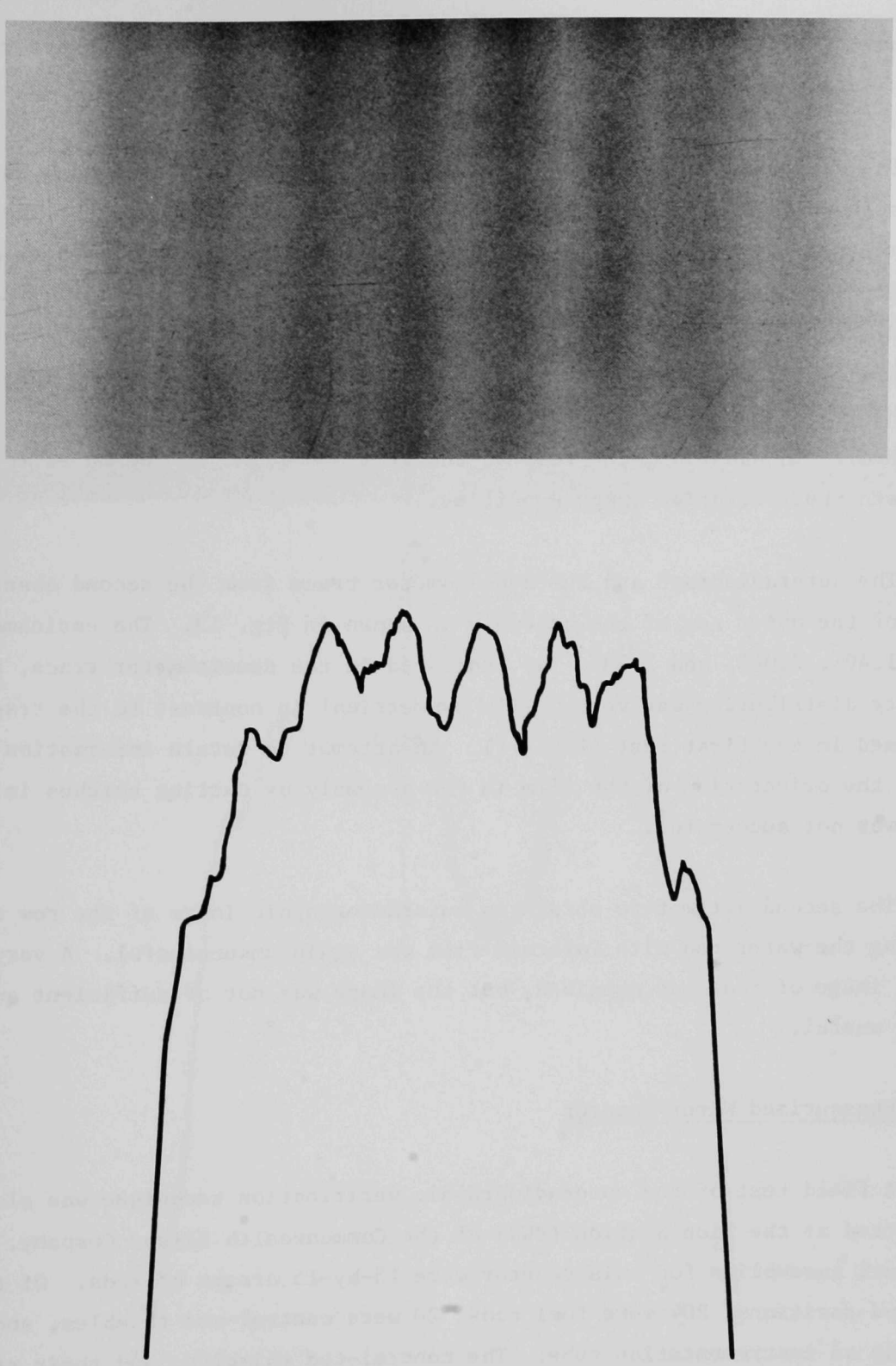


Fig. 11. Autoradiograph and scanning-densitometer trace of a row of rods in a BWR fuel assembly. Ortho-G film, Lanex screens, and rod-isolation shielding. ANL Neg. No. 150-78-20.

All cassettes and shielding were supported by the rod spacers. Exposure time was 3 hours.

The second autoradiograph of the assembly row which contained a water rod is shown in Fig. 12, along with a scanning-densitometer trace. The trace is similar to that obtained in the first test, but there are also some important differences. In the first test, the rod on the left end (2.14% enriched) had an image density less than that of the rod on the right end (2.87% enriched). In the second test, the image of the rod on the left end (2.06% enriched) had a greater density than that of the rod on the right end (2.73% enriched). In addition, the rods to the left and right of the water rod reversed their relative image densities.

The autoradiograph and the densitometer trace from the second observation of the outer row of the assembly is shown in Fig. 13. The enrichments were 1.40%, 2.06%, and 2.73%. As indicated in the densitometer trace, the density distribution was very nearly symmetrical in contrast to the trace obtained in the first test (Fig. 11). An attempt to retain information about the orientation of the film in the assembly by cutting notches in the film was not successful.

The second attempt to obtain an autoradiographic image of the row containing the water rod with Polaroid film was again unsuccessful. A very faint image of rods was obtained, but the image was not of sufficient quality to be useful.

## B. Pressurized Water Reactor

A field test of the autoradiographic verification technique was also conducted at the Zion Station (PWR) of the Commonwealth Edison Company. The fuel assemblies for this reactor were 15-by-15 arrays of rods. Of the 225 rod positions, 204 were fuel rods, 20 were control-rod thimbles, and one was an instrumentation tube. The control-rod thimbles, and their effect on obtaining autoradiographs, were discussed in Section III-C. The instrumentation tube was located in the center of the assembly and had the same diameter as the control-rod thimbles. All fuel rods contained 2.80% U-235.

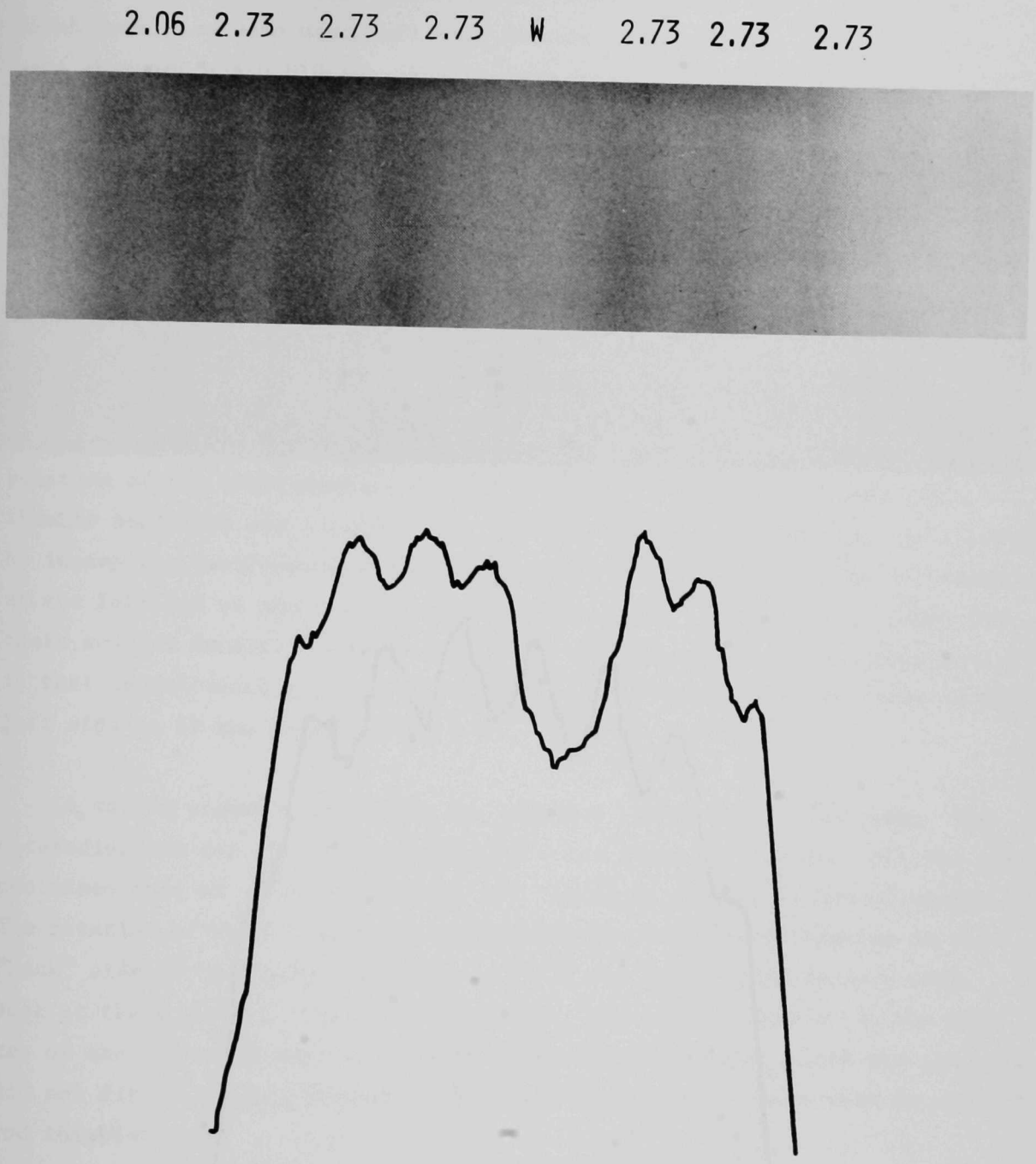



Fig. 12. Autoradiograph and scanning-densitometer trace of a row of rods in a BWR fuel assembly. Ortho-G film, Lanex screens, and row-isolation shielding. Rod W contains no uranium. ANL Neg. No. 150-78-24.

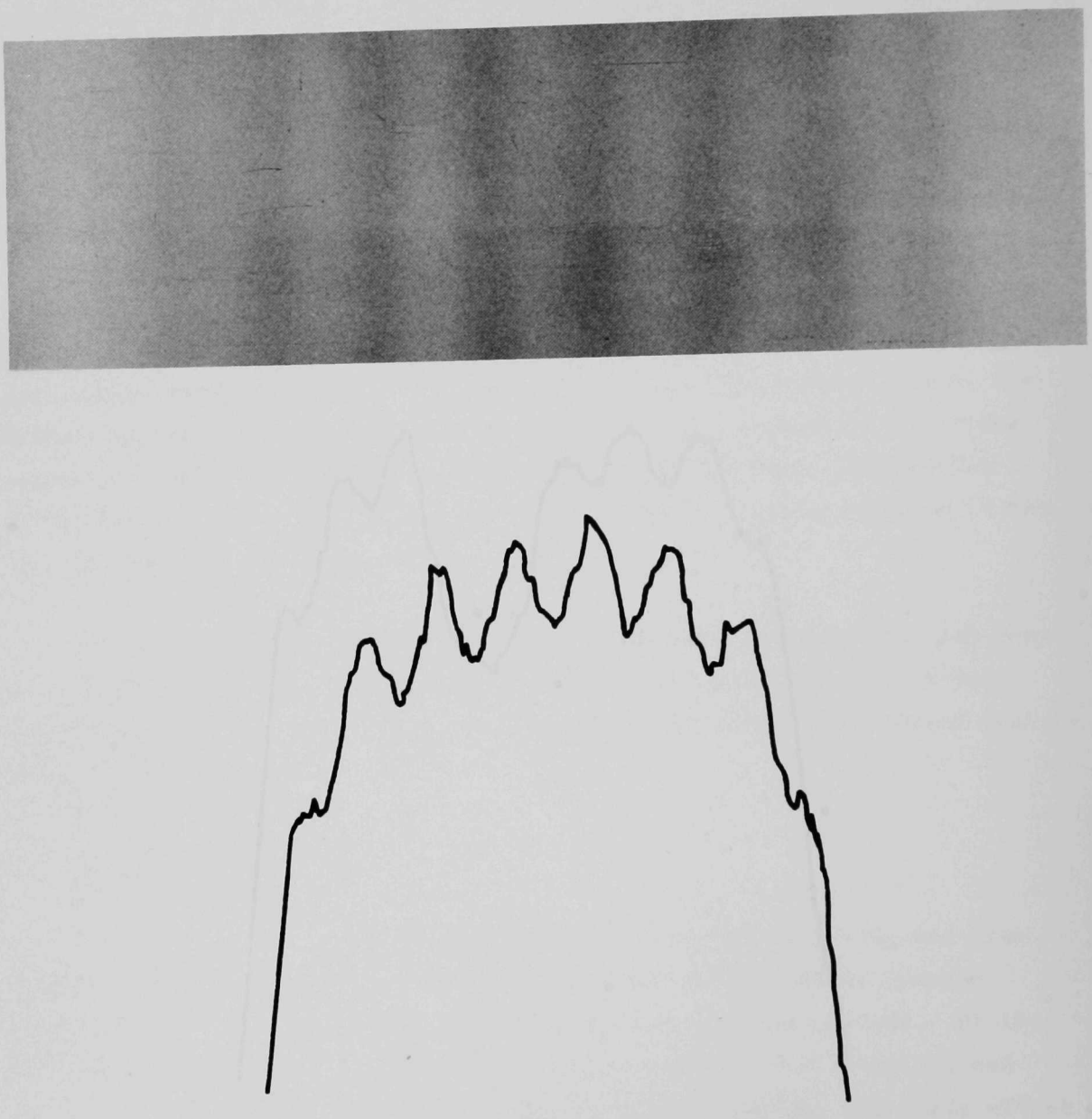


Fig. 13. Autoradiograph and scanning-densitometer trace of a row of rods in a BWR fuel assembly. Ortho-G film, Lanex screens, and row-isolation shielding. ANL Neg. No. 150-78-15.

The autoradiographic test was conducted in the fuel-storage vault. Assemblies were held vertically in metal racks, and the rack structure prevented film access to the three outer rows of the assembly. Film cassette composition was similar to that of the second Dresden test. Four 70-mm-wide Kodak envelopes were used, all with Ortho-G film and Lanex screens. The beta absorber was 0.010-in. of stainless steel, and the gamma absorber was 0.022 in. of lead. The shielding was 0.055 in. of lead. Again, the cassettes and the shielding were enclosed in polyethylene envelopes. Total cassette thickness was approximately 0.100 in., and the test exposures were made at the lower part of the assembly where the control-rod thimbles had a diameter of 0.489 in. Row-isolation shielding was placed parallel to the film. Exposure was for 3 hours.

The center row of the assembly was one of the rows examined. The autoradiograph and its scanning-densitometer trace are shown in Fig. 14. The position of the instrumentation tube is labeled "I," and the control-rod thimble positions are labeled "C." The rod image on the far left is obscured by incomplete development at that end of the film. The other two rod images at the left end of the trace were fully developed. One possible reason for their reduced density compared to that of the rod images on the right side is that cassette-rod contact, or film-screen contact was not as close on the left side as it was on the right side.

A second assembly row which was examined contained 15 fuel rods. The autoradiograph and the densitometer trace are shown in Fig. 15. All the rods contained fuel of equal enrichment, but the image density is not constant. The location of maximum density corresponds to control-rod thimbles on the "back" side of the film. Cassette-fuel rod and film-screen contact were best at these points of minimum clearance. The reduced density in the center of the row could be due to poorer film-screen contact, since the cassette did not fit as tightly between rows in the center where there were no control-rod thimbles.

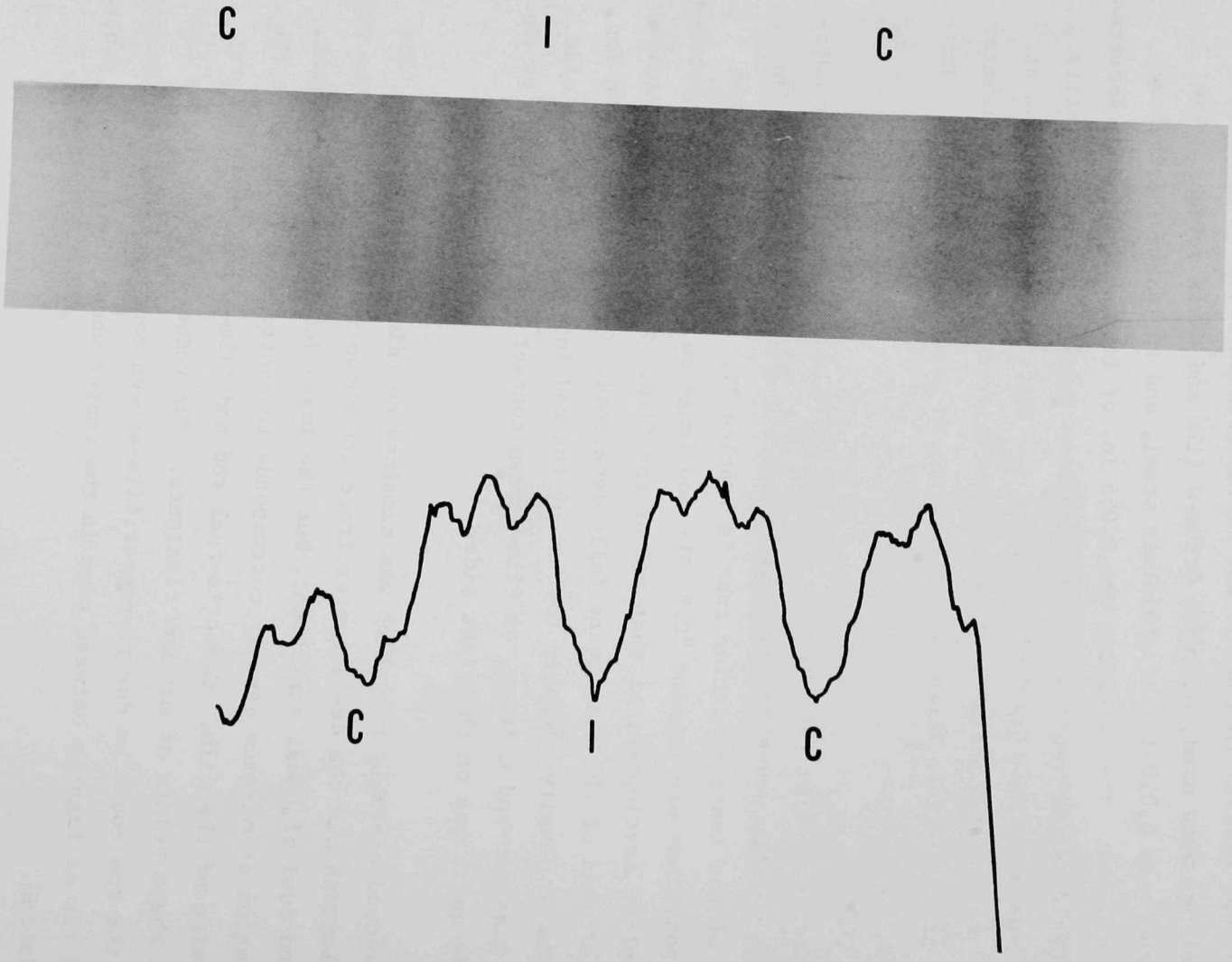



Fig. 14. Autoradiograph and scanning-densitometer trace of a row of rods in a PWR fuel assembly. Ortho-G film, Lanex screens, and row-isolation shielding. Rods I and C contain no uranium. ANL Neg. No. 150-78-23.

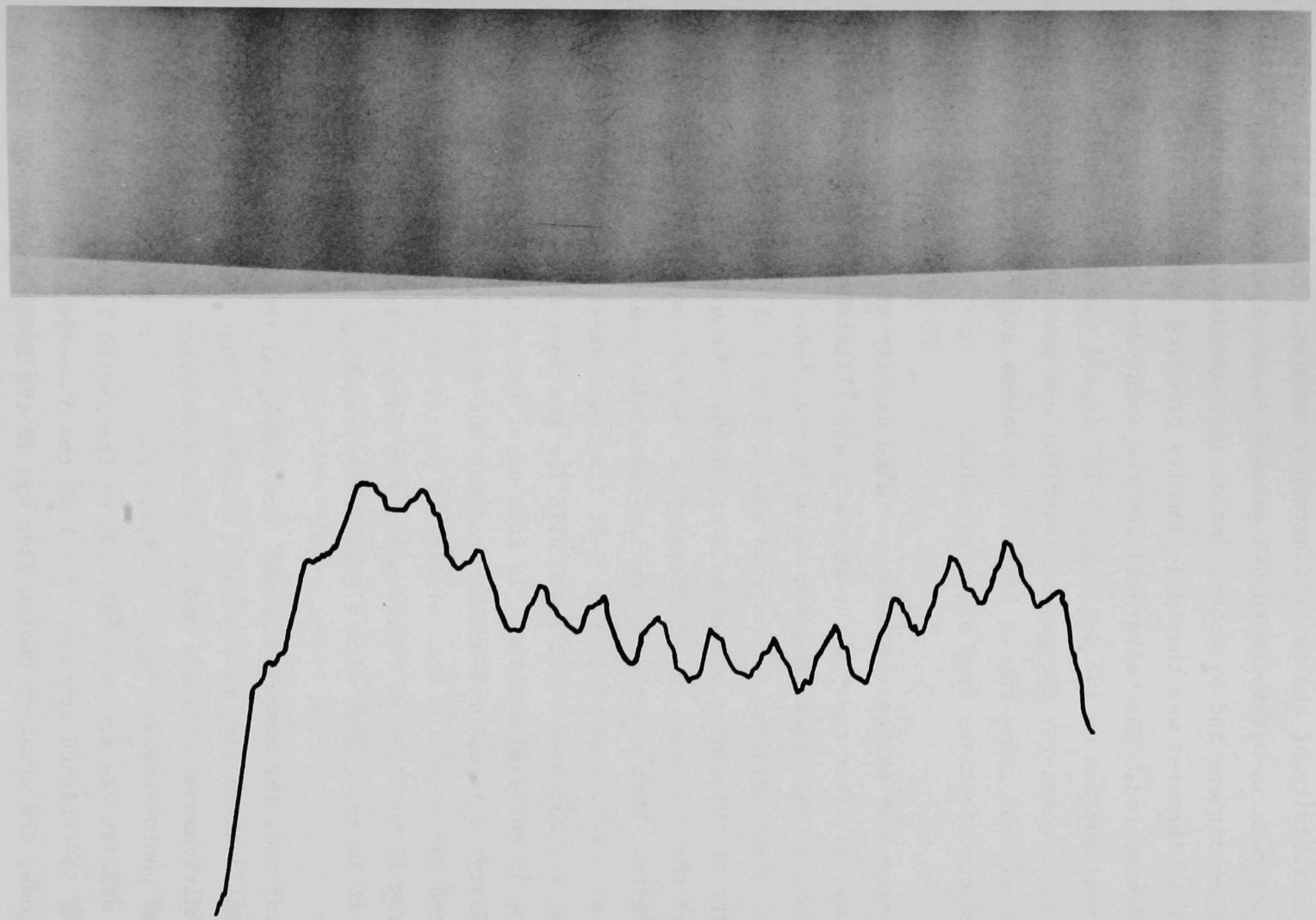


Fig. 15. Autoradiograph and scanning-densitometer trace of a row of rods in a PWR fuel assembly. Ortho-G film, Lanex screens, and row-isolation shielding. ANL Neg. No. 150-78-16.

# V. DISCUSSION

The sensitivity of the autoradiographic technique to the U-235 enrichment of fuel rods in unirradiated LWR assemblies was limited by signal-to-background conditions and by uncertainties in image-density measurements. The signal of interest was the image density produced by the U-235 contained in a particular rod. The background was the image density produced by the U-238 in that rod and by the U-238 and U-235 in all the other rods in the assembly. The gamma-ray background contributions were only partially reduced by the use of lead gamma-ray shielding. Stainless steel shielding virtually eliminated contributions from beta radiation.

The extent of the contribution to image density at a particular rod position by the other rods in the assembly was illustrated by densitometer measurements of the autoradiograph shown in Fig. 6-A. Here, the density of the rod image adjacent to the void was 0.77 optical-density units, while the density at the center of the void was 0.60. The difference between the density of the rod image and the density of the void was only 0.17 optical-density units. When a depleted-uranium-containing rod occupied the central position of a row, such as in Fig. 7-B, its image density was 0.71 optical-density units, compared to 0.77 and 0.78 for the images of the adjacent rods containing 3%-enriched uranium. In this case, the difference between the image density of a depleted-uranium rod and the image density of an enriched-uranium rod was only 0.07 optical-density units. Even though an enrichment change from 1% to 2% U-235 represents a very large relative change in U-235 content, it is associated with a small relative change in image density.

In any rod, the gamma radiation from U-238 and its daughters made a substantial contribution to the image density of that rod. Measurements using a multichannel analyzer and an NaI(Tl) detector showed that, in the 66-283 KeV photon-energy range, the total photon flux from a rod containing depleted uranium was approximately 65% of the photon flux from from a rod containing 3%-enriched uranium. For both the 3%-enriched and the depleted-uranium rods, the greatest photon flux was in the X-ray energy range at approximately 100 KeV. The 66-283 KeV energy range was chosen because the major gamma rays from U-235 were included and the film had high sensitivity

in this range. The relatively large contribution from U-238 and its daughters to the total low-energy photon flux, combined with contributions from other rods, was the probable cause for the small density difference observed between the image of a depleted-uranium-containing rod and the image of a 3%-enriched uranium-containing rod.

Because determinations of changes in enrichment were dependent upon measuring relatively small changes in image densities, it was necessary to estimate the uncertainties in density-difference measurements. Measurement uncertainties were a major factor in the assessment of the enrichment sensitivity of the autoradiographic technique. The results of the field tests at reactor sites indicated significant variations in image density between different rods located in equivalent positions in the assembly and containing equivalent U-235 concentrations. A further experiment using the model assembly was conducted to establish the degree of variation in image density and in density profiles for uniform enrichment. Autoradiographs were obtained with cassettes whose thickness was slightly less than the rod spacing. Eight separate exposures were obtained from rows of rods containing uranium of equal enrichment under conditions as nearly identical as possible. The resulting densitometer scans revealed density and density-profile variations as great as those between the scans in Figs. 10 and 12 and in Figs. 11 and 13. Absolute-density measurements were made with a spot densitometer. These measurements showed a standard deviation of 0.012 optical-density units in the difference between the image density of the central rod and the image density of the gap between the central rod and the adjacent rod. These density differences were approximately 0.06 optical-density units and were divided by the central-rod image density to remove variations due to film-processing conditions. The standard deviation of 0.012 optical-density units is about 18% of the density difference between the image of a depleted-uranium-containing rod and the image of a 3%-enriched uranium containing rod. At a 95% confidence level (2 standard deviations), the uncertainty is 36% of the enriched rod-depleted rod density difference. This means that enrichment could be determined only to $3 \pm 1$ %. A rod enriched to 2% U-235 would have overlapping confidence intervals with a 3%-enriched rod, and this enrichment difference could not be reliably detected.

Better enrichment sensitivity for U-235 has not been gained from autoradiography because film detectors have very poor energy resolution. The main difference between the photon spectra from U-235 and from U-238 is the presence of the 185 KeV peak present only in U-235. However, both U-235 and U-238 emit photons at other energies, mostly X-rays, which effectively contribute to film density. The NaI(Tl) measurements indicated that 65% of the photons with energy between 66 and 283 KeV from a 3%-enriched rod were from U-238, and 35% were from U-235. When optical-density measurements were made on the autoradiographs of 3%-enriched rods in the model assembly, the density was 0.77, compared to 0.71 for a depleted rod, but the density at the position of a void was 0.60. If the density of 0.60 for the void is subtracted as a background correction, the remaining densities for the 3%-enriched and the depleted rods are 0.17 and 0.11, respectively. The difference of 0.06 for the 3% rod is then 35% of the total, and the 0.11 for the depleted rod is 65% of the total, which agrees with the NaI measurements. This indicates that if a NaI(Tl) system were used to measure only gross counts integrated over a wide energy range, as is the case with film, then the film and the NaI(Tl) detector would have equivalent sensitivity to enrichment.

From these observations it was concluded that the small changes in image densities or in image-density profiles, such as those observed in Figs. 10-13, were within the uncertainties of the measurement and could not be related to small changes in enrichment. It was also concluded that the small enrichment changes encountered within a row of rods in typical BWR assemblies could not be reliably detected by this technique.

One possible explanation for the difference in image density for equivalent rods is nonuniform contact between film and screens. It is well known in radiography that tight, uniform contact between film and screens is necessary for uniform density and for good image quality. For this reason, medical X-ray film cassettes incorporating screens are backed by sponge rubber to keep a constant, uniform pressure on the film and screens. For the thin, flexible cassettes used in these studies, the pressure to keep screens and film in close contact had to come from the rods in the assembly. However, to ensure that the rods would not be deformed, the reactor operators did not permit the fuel rods to be placed under stress. Perhaps the strongest evi-

dence suggesting the relationship between film-screen contact and image density is the autoradiograph in Fig. 15.

Poor film-screen contact is also a likely reason for the relatively unsatisfactory quality of the autoradiographic images obtained with Polaroid film. Polaroid film was attractive for this application because of its simple processing requirements, but it is thinner and less resilient than medical X-ray film, and this may have caused less even contact. Polaroid images appeared to be similar to other types of X-ray images where film-screen contact was poor.

The field tests at the Dresden reactor (BWR) were conducted with the assemblies in the test stand, as reported in Section IV-A. This test stand was the only location in the reactor station where a fuel assembly could be placed to make it accessible to autoradiographic examination. The fuel-storage vault did not offer sufficient accessibility. The test stand only accommodated two assemblies, and fuel-assembly handling was kept to a minimum, so there was no opportunity for examining a large number of assemblies in a short time at Dresden. Because international inspectors need to examine many assemblies in a short time, a facility such as Dresden would present serious problems for a large-scale application of autoradiographic inventory techniques. In contrast to Dresden, the Zion Station fuel-storage vault provided ready access to the fuel assemblies, and should present no problems for a large-scale inventory. Other facilities where autoradiography might be applied are those which fabricate LWR assemblies.

An important observation resulting from the studies of image-density dependence on the U-235 content of single rods was the contribution made by U-238 and U-238 daughters to image density when insufficient filtration was used. Part of this image-density contribution was attributed to the beta radiation from Pa-234m. The beta-radiation contribution was greatly reduced by adding stainless steel absorber which had a high cross section for beta absorption but which had a low cross section for gamma absorption. The reversal of the sign of the slope of the line for image density versus U-235 concentration upon adding stainless steel absorber was considered to be evidence that part of the contributions from U-238 were caused by beta radiation.

The incorporation of beta absorber in film cassettes is important because it reduces the possibility of undetected diversion by the simultaneous substitution of depleted uranium for enriched uranium and the alteration of the beta absorption properties of the fuel-rod cladding. Possible cladding alterations are substitutions of less dense or thinner material.

The autoradiographic technique described here can provide only a relative indication of the U-235 content of rods in an assembly. It is not an absolute-enrichment measurement. An autoradiographic inventory needs to be supported by quantitative, absolute-enrichment measurements on the exterior rods of the assemblies tested. Once the absolute enrichment of exterior rods is established, autoradiographs can verify enrichment consistency throughout the assembly. The use of an absolute-enrichment measurement would help to detect diversion attempted by the substituting of depleted-uranium-containing rods for all rods in an assembly. The autoradiographic method could possibly be made more quantitative by including an exposure standard which produces a reference film density during a verification exposure.

Most difficult was the detection of depleted-uranium-containing rods at all interior-rod positions in a row when the end-rod positions in that row were occupied by 3%-enriched uranium-containing rods. This defect was usually detectable, but only with very careful examination, and the difference between a defect-containing row and a normal row was very subtle. The defect was easier to detect with densitometer traces. Measurements with a spot densitometer were also helpful. It may possibly be beneficial to try to make absolutement-enrichment measurements at the gap between rods as well as at the rod centers to try to detect this defect.

Despite the problems attributed to nonuniform film-screen contact and to high levels of background radiation, the autoradiographic technique was consistently able to detect dummy rods, missing rods, or rods containing depleted uranium. These defects could be detected by visual inspection of processed film. The technique has the great advantage of being simple to apply, of requiring very little equipment, and of requiring no sophisticated instrumentation. The technique was acceptable to the utility operating the reactors at which the field tests were performed. All items necessary for an auto-

radiographic inspection are easily portable. Access to at least a dark chamber is necessary if film is to be processed in the facility under inspection. Cassettes can be loaded in advance of an inspection. Both Ortho-G and XR medical X-ray film can be processed either in automatic processing equipment or by hand with very simple equipment and standard developing and fixing solutions. Even the Polaroid film requires a dark chamber, because the film must be removed from its packet for exposure and reinserted for processing. Many autoradiographs can be exposed simultaneously, and the film subsequently batch processed, which allows many assemblies to be sampled.

## VI. CONCLUSIONS

It has been shown that autoradiography provides a means of verifying that rods in the interior of an unirradiated LWR fuel assembly do contain U-235. The technique provides a relative indication of U-235 content and must be accompanied by an absolute-enrichment measurement for external rods. Since interior rods in assemblies are not subject to verification under present international inspection procedures, the autoradiographic technique may prove useful in providing more effective safeguards of the U-235 contained in fuel assemblies deployed internationally. The autoradiographic technique has the advantage of being simple to implement, and is capable of detecting defects such as dummy rods, missing rods, and rods containing depleted uranium. These defects can be detected by visual inspection of the autoradiographs.

ACKNOWLEDGEMENT

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## Distribution for ANL-78-27 and ISPO-12

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